

PCT

WORLD INTELLECTUAL PROPERTY ORGANIZATION
International Bureau

INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification ⁵ : G11C 13/04	A1	(11) International Publication Number: WO 93/02454 (43) International Publication Date: 4 February 1993 (04.02.93)
(21) International Application Number: PCT/US92/05858 (22) International Filing Date: 22 July 1992 (22.07.92) (30) Priority data: 733,030 22 July 1991 (22.07.91) US (71) Applicant: CORNELL RESEARCH FOUNDATION, INC. [US/US]; Cornell Business and Technology Park, 20 Thornwood Drive, Suite 105, Ithaca, NY 14850 (US). (72) Inventors: STRICKLER, James, H. ; 210 Clark Hall, Ithaca, NY 14850 (US). WEBB, Watt, W. ; 409 Highland Road, Ithaca, NY 14850 (US).		(74) Agents: COOPER, George, M. et al.; Jones, Tullar & Cooper, Suite 1002, 2001 Jefferson Davis Highway, Arlington, VA 22202 (US). (81) Designated States: JP, European patent (AT, BE, CH, DE, DK, ES, FR, GB, GR, IT, LU, MC, NL, SE). Published <i>With international search report.</i>
<p>(54) Title: METHOD FOR THREE-DIMENSIONAL OPTICAL DATA STORAGE AND RETRIEVAL</p> <p>(57) Abstract</p> <p>In a method of high density (superior to 10^{12} bits/cc) optical recording of information in a three-dimensional multilayered format, information is written in a photopolymer (10) as submicron volume elements of altered index of refraction. The index change results from alteration of the photopolymer (10) induced by two-photon excitation of the photo-sensitizer at the waist of a highly focused beam (20) from a subpicosecond colliding pulse modelocked laser (16). Quadratic dependence of two-photon excitation on incident instantaneous intensity serves to confine crosslinking of the polymer (10) to the focal volume; hence, bit array layers may be written with thicknesses on the order of the confocal parameter. As an example, eight well resolved bit planes can be written in a polymer film about 30 microns thick. Information written in this manner may be read with sufficient axial resolution by differential interference contrast (DIC) or confocal microscopy. This technique may be employed in multilayered write once read many (WORM) optical storage discs, for example.</p>		

FOR THE PURPOSES OF INFORMATION ONLY

Codes used to identify States party to the PCT on the front pages of pamphlets publishing international applications under the PCT.

AT	Austria	FI	Finland	ML	Mali
AU	Australia	FR	France	MN	Mongolia
BB	Barbados	GA	Gabon	MR	Mauritania
BE	Belgium	GB	United Kingdom	MW	Malawi
BF	Burkina Faso	GN	Guinea	NL	Netherlands
BG	Bulgaria	GR	Greece	NO	Norway
BJ	Benin	HU	Hungary	PL	Poland
BR	Brazil	IE	Ireland	RO	Romania
CA	Canada	IT	Italy	RU	Russian Federation
CF	Central African Republic	JP	Japan	SD	Sudan
CG	Congo	KP	Democratic People's Republic of Korea	SE	Sweden
CH	Switzerland	KR	Republic of Korea	SN	Senegal
CI	Côte d'Ivoire	LJ	Liechtenstein	SU	Soviet Union
CM	Cameroun	LK	Sri Lanka	TD	Chad
CZ	Czechoslovakia	LU	Luxembourg	TC	Togo
DE	Germany	MC	Monaco	US	United States of America
DK	Denmark	MG	Madagascar		
ES	Spain				

METHOD FOR THREE DIMENSIONAL OPTICAL

DATA STORAGE AND RETRIEVALBackground of the Invention

5 This invention was made with Government support under Grant No. P41RR04224 awarded by the National Institutes of Health, and under Grant No. DIR8800278, awarded by the National Science Foundation. The Government has certain rights in the invention.

10 The present invention relates, in general, to a method of storing data in a three-dimensional medium, and more particularly to optical data storage techniques utilizing two-photon excitation in refractive media.

15 Conventional two-dimensional optical data storage can register information at about 10^8 bits/cm² using visible or infrared wavelengths at the diffraction limit. It has been proposed that by writing and reading data in a three-dimensional
20 format, data storage densities of greater than 10^{12} bits/cm³ could be achieved. See, for example, U.S. Patents Nos. 4,466,080 and 4,471,470 to Swainson et al, which disclose the use of two intersecting beams of radiation which are matched
25 to selected optical properties of an active medium to form and to detect inhomogeneities. In such a system, a stack of two-dimensional (2-D) planar bit arrays effectively multiplies data density by the number of planes in a three-
30 dimensional (3-D) stack. Although the chief difficulty with such a scheme is cross talk between planes, writing with three dimensional resolution in thick media can be accomplished by

non-linear excitation of the media to confine data storage to the selected focal plane. To see how this can be done, consider a single focused Gaussian beam, well below saturating intensity, incident on a physically thick but optically thin absorbing sample. For the case of excitation that is linear in the direction of the incident radiation, the same amount of energy is absorbed in each plane transverse to the optical axis regardless of distance from the focal plane, since nearly the same net photon flux crosses each plane. Thus, linear excitation strongly contaminates planes above and below the particular focal plane being addressed. For an excitation scheme which depends quadratically on the intensity, however, net excitation per plane falls off with the inverse of the square of the distance from the focal plane. Therefore, information can be written in a particular focal plane without significantly contaminating adjacent planes beyond the Rayleigh range $Z_0 = \pi w_0^2 n / \lambda$, where n is the medium index of refraction, λ is the wavelength, and $w_0 = 1.22 \lambda / \text{N.A.}$ The minimum spot size may be approximated by the Rayleigh criterion for a round aperture.

Patents Nos. 4,466,080 and 4,471,470 proposed the use of two or more intersecting beams to localize the writing and reading of information on 3-D photochromic optical memories. Patent No. 5,034,613 to Denk et al, the disclosure of which is incorporated herein by reference, proposed a simpler scheme employing a single highly focused beam to record and read via two-photon excitation.

Subsequently, the applicants herein demonstrated high density optical data storage via two-photon photoactivation of a fluorescent dye which was non-fluorescent until photochemically modified, as described in "Two-photon Excitation in Laser Scanning Fluorescence Microscopy", Proceedings of the International Society for Optical Engineering, Vol. 1398, pp. 107-117, 1991. However, memories which rely on fluorescence modulation suffer the limitation that their useful lifetime is limited by photobleaching.

Summary of the Invention

The foregoing problems are overcome, in accordance with the present invention, through the provision of optical techniques for writing and reading data in a three dimensional multilayered format wherein the information is written as submicron voxels of modified index of refraction that are induced by two-photon excited crosslinking or degradation of polymer media in the volume defined by the waist of a highly focused modelocked laser. Information so stored is later read with three dimensional resolution via laser differential interference contrast microscopy. These techniques allow storage of data at an unprecedented density of greater than 10^{12} bits/cc., and their application to a spinning disc format has the potential to produce memory devices having 100 times greater information storage capacity than currently available 2-D disc machines. The method also appears suitable for the fabrication of computer generated volume

holograms of the type which have been proposed for important applications in optical computing.

Two-photon excitation refers to the simultaneous absorption of two photons by a chromophore molecule. Frequently, real excited states which are normally accessed via single photon absorption may also be excited via absorption of two quanta each having half the energy of the single photon. The essential characteristic of the process in the present context is that the photons must simultaneously impinge on the molecule; thus, the excitation rate is proportional to the square of the incident intensity. Excitation is thereby confined to the ellipsoidal focal volume where the intensity is extremely high. Such excitation is produced, in the preferred form of the invention, by a laser which provides sufficient incident intensity to produce simultaneous absorption of two photons by the photopolymer molecules, each photon having one-half the energy required for normal single photon absorption.

Brief Description of the Drawings

The foregoing, and additional objects, features and advantages of the present invention will become apparent to those of skill in the art upon a reading of the following detailed description of a preferred embodiment thereof, taken in conjunction with the accompanying drawings, in which:

Fig. 1 is a diagrammatic illustration of experimental apparatus for writing and reading

three dimensional refractile optical memories, in accordance with the invention;

5 Figs. 2 and 3 are images of adjacent random bit planes taken from a stack after they were written as a three-dimensional optical memory by the apparatus of Fig. 1;

Fig. 4 is an image of a longitudinal cross-section through a stack of 10 of the planes of Figs. 2 and 3; and

10 Fig. 5 is an image of a longitudinal cross-section through the central region of a 25 layer memory stack.

Description of Preferred Embodiment

Two-photon photo-initiation of a
15 crosslinking reaction in a material such as polymer which leads to a density increase of the material and concomitant increase in the index of refraction may be used to write phase data in three dimensions. Applicants have found that a
20 liquid acrylate ester blend such as CIBATOOL XB5081, available from 3d Systems, Valencia CA, may be solidified by two-photon excitation using highly focused high peak power pulsed laser irradiation of 620 nm wavelength. Upon exposure,
25 solidified regions of this material become visible within the liquid due to refraction, suggesting this sort of material as a candidate for an optical memory medium. Many photoresists are known to undergo density changes upon either
30 crosslinking or photodegradation and should also be useful as phase recording memory media. Furthermore, reversible photorefractive materials may potentially be used as erasable optical

memories. Such materials have already been used in holographic three dimensional data recording schemes but only with considerably lower data storage density.

5 As with the case of fluorescent memories, refractile memories must be read via a technique which has axial resolution as high as the writing method and which discriminates against an out of focus background. Differential
10 interference contrast microscopy (DIC), which is a well-known technique of microscopy, may be made to be sensitive to optical path differences originating only at the focal plane. This is accomplished by using laser beams to scan the
15 material, instead of illuminating the full field, so that two focused cross polarized components of a plane polarized source beam are made to pass through the specimen separated by a small distance ($<1\mu\text{m}$) in the transverse direction. At the focal
20 plane, where the component beams are closely adjacent, or overlap by only a small amount, they will suffer a relative phase shift if the two beams pass through regions of different refractive indices. By detecting and then recombining these
25 beams so that they interfere in the image plane, and by analyzing the polarization of the detected beams relative to the source beam polarization, it is possible to measure the relative phase shift that occurred between the two beams within the
30 material at the focal plane. By raster scanning the focused beams through the material and displaying the detected intensity on a CRT, an image of the transverse gradient of the index of

refraction at the focal plane is produced. Since the component beams are largely overlapping outside of the focal plane this technique is sensitive to information only at the focal plane.

5 Phase sensitivity can be further improved by recording two orthogonal polarizations and computing a normalized differential signal. (See, for example, W. Denk and W.W. Webb, Applied Optics, Vol. 29, p. 2382 (1990)).

10 An optical memory in accordance with the present invention may be in the form of a film of the photopolymer CIBATOOL, which may be prepared for data storage as an approximately 100 μ m thick film sample placed between a microscope slide and
15 a coverslip. The film sample is first irradiated with about 3 mJ/cm² of UV light in order to gel the sample and thereby prevent distortion due to shrinkage and flow. Sample 10 is then placed on the stage 11 of an inverted microscope generally
20 indicated at 12, and which may be a Zeiss Model IM-35. The sample is irradiated for data storage through a high numerical aperture (N.A.) objective lines 14, such as a Nikon planapo 60X 1.4 N.A., with 100 fs pulses of, for example, 620 nm
25 wavelength light from a colliding pulse modelocked dye laser diagrammatically illustrated at 16, pumped by an Argon ion laser 18. Laser beam 10 is supplied to microscope 20 by way of dichroic mirror 21 and computer controlled scanning mirrors
30 (not shown) of a laser scanning confocal microscope 22 such as model MRC-600 from Bio-rad. The sample 10 carried on stage 11 is translated in the axial, or Z, direction by a stepper motor

focus controller 24 which is connected to the focusing knob 26 of the microscope 12, and which is controlled by a computer 28. The computer 28 is also connected to the scanning mirrors of microscope 22 to control the X-Y translation of the irradiating beam 20 from laser 16 on the sample.

By moving the laser beam 20 in the X-Y direction, a two-dimensional bit plane is defined, with transverse locations being scanned by the beam 20 to define data points in that plane. By moving the objective lens 14 in the Z direction so as to focus at different planes, different two-dimensional bit planes are defined so that three-dimensional stacks of data can be written into the sample. Writing occurs when the light focused at a location in the sample has sufficient instantaneous intensity to modify the material of the sample, as by producing two-photon photopolymerization of the sample material. Such a polymerization produces a change in a characteristic of the specimen material, such as a change in its refractive index, at a very small point, or pixel, in the material. This change produces a bead of material which is different than surrounding material outside the focal point of the scanning beam and thus outside the region of two-photon excitation of the sample material, and this change may be referred to as a data bit at that focal point, or pixel location.

In accordance with experiments utilizing the apparatus of Fig. 1, the X-Y scanning and the Z-direction focus is shifted by the computer 28

periodically in a predetermined pattern to provide a dwell time for the scanning beam of about 10 ms at each selected pixel location. The dye laser 16 produces intense 100fs pulses of light at the repetition rate of 100 MHz to produce 2-3 mW average power. Exposure of the material at each selected pixel location to this light for a period of about 10ms produces a three-dimensional array of distinct, nonoverlapping optically refractive beads at the selected locations in the specimen. This array is in the form of stacked layers of X-Y data bit planes, with the refractive beads representing data bits.

Optically refractive beads formed in an array in accordance with the foregoing are illustrated in Figs. 2 and 3, wherein a pair of adjacent bit planes 34 and 36 are illustrated as including multiplicities of pixel locations 38. The planes 34 and 36 were adjacent to each other in a stack, with the adjacent planes being separated by about 3 micrometers. The pixel locations in each plane are separated in transverse directions by about 1 micrometer, center to center, producing a bit density of 0.3×10^{12} bits/cm³ in the stack. Data written into pixel locations on the two planes are indicated by grey regions 40.

A longitudinal cross-section through a stack of 10 planes, generally indicated at 42-51, is illustrated in Fig. 4. The planes in this stack are separated in the Z direction by 3 micrometers, as noted above. Similar structures up to 30 planes thick, with data densities of 1.6

10

x 10^{12} bits/cm³, have been generated, with Fig. 5 showing a section from a 25-layer structure, having a density of 1.3×10^{12} bits/cm³. It is noted that in the image of Fig. 4, a second order background subtraction has been digitally performed to remove the field ramp produced by the DIC imaging method.

The optical memory formed by the foregoing process may be read by successively imaging each of the data-containing planes, such as planes 42-51, by directing laser light into the sample 10 carried by stage 11, again using the optics of the laser scanning microscope 22. The presence or absence of optically refractive beads, at specified X,Y and Z pixel locations in the sample can then be determined, so as to retrieve the data previously written. More particularly, an Argon ion laser 50 (Fig. 1) directs a read beam 51 of light at a wavelength of 488 nm, for example, onto the specimen by way of dichroic mirror 21, the scanning mirrors of the laser confocal microscope 22, and the optics of the inverted microscope 12. The read beam 51 passes through the specimen and through lens 52 to a suitable light pipe 54 which directs the light to a photomultiplier light detector which forms a part of laser scanning microscope 22. The read beam scans each layer of the stack in the X-Y plane to produce a corresponding image, and by successively focusing the microscope 12 on the various planes by way of focus control 24, each layer of the stack can be read. The plane images so produced may be displayed on monitor 56 and may

be supplied to suitable processing circuitry to determine the locations of those pixels which include data bits, for example in the form of refractory beads formed by the data writing process described above.

As discussed above, to read, or image, the closely spaced bit planes of a sample, differential interference contrast (DIC) microscopy, as described by G. Nomarski, Journal Phys Radium 16, 9s-11s (1955), is used, since it is sensitive to optical path differences originating only at the focal plane in the sample. The laser scanning microscope converts the plane polarized read beam 51 into two focussed cross polarized components which pass through the sample 10 separated by a small distance (less than 1 micrometer) in the transverse direction. At the focal plane in the specimen, where the component beams barely overlap, they will undergo a relative phase shift, caused by one of the beams passing through a pixel, or data bit region, where the index of refraction has changed while the other beam passes through an adjacent region that is unchanged. After passing through the sample, these beams are recombined so that they display an interference pattern in the image plane. By analyzing the polarization of the image beams with respect to the source polarization, it is possible to measure the relative phase shift produced in the beams at the focal plane.

By scanning the focused beams across each layer of the sample in sequence, an image of

the transverse gradient of the index of refraction for a stack of data planes can be produced.

Three-dimensional refractile data storage is entirely adaptable to the rotating disc format used in current commercial optical data storage products. A reading device requires incorporation of DIC transmission optics, and a recording (or writing) device requires a high peak power ultrafast modelocked laser source or other convenient short pulse source. Alternatively, resonance enhanced two photon absorbing media may provide much higher two-photon cross sections that could allow recording with a conventional laser diode.

Because the volume of the written bit depends on $1/(N.A.)^4$, the use of high numerical aperture focusing optics is essential to maximize data density. While using a nonimmersion objective with, for example, $N.A. = 0.9$ may reduce the data density by a factor of 6 relative to the best oil immersion objectives, the theoretical limit remains at about 10^{12} bits/cm³. Furthermore, since the number of layers that may be written depends on the lens working distance, dry lenses should allow use of a thicker memory structure with more data layers.

While the writing speed of the experimental system described above is relatively slow, an increase of the power of the incident beam by a factor of 100 using currently available lasers would increase the writing speed for this material by 10,000. By judicious optimization of material and irradiation wavelength for high two-

13

photon absorption cross section, and by use of higher incident power, submicrosecond voxel writing times are anticipated. The writing time may ultimately be limited only by the repetition rate of the pulsed laser, while the maximum reading speed for the 3-D optical memory should be limited only by the mechanics of the scanning system and the spinning disc, as in current 2-D systems.

10 Although the present invention has been described in terms of a preferred embodiment, it will be understood that variations and modifications may be made without departing from the true spirit and scope thereof, as set forth in
15 the following claims.

WHAT IS CLAIMED IS:

1. A method for writing optical data with three-dimensional resolution, comprising:
producing a single, intense beam of focusable, coherent light;

focussing the beam on a photosensitive material to produce two-photon excitation of said material at the focal point of the beam to thereby produce a characteristic change in said material.

2. The method of claim 1, wherein the focussing of said beam produces a refractive index inhomogeneity in said material at said focal point.

3. The method of claim 1, wherein the focussing of said beam produces a refractive index inhomogeneity by modification of the density of the photosensitive material.

4. The method of claim 1, further including:

scanning said beam through said material to produce characteristic changes in said medium at selected locations to thereby produce three-dimensional inhomogeneities in said medium.

5. The method of claim 4, wherein the step of scanning said beam includes moving a focal point of said beam in an X-Y plane within said material to define a plurality of pixel locations in said X-Y plane.

6. The method of claim 5, wherein the step of scanning said beam further includes shifting the focal point of said beam along a Z axis to define a plurality of X-Y planes within said material, to thereby define a three

dimensional array of pixel locations in said material.

7. A method of reading optical data stored as a refractive index inhomogeneity in a medium, comprising:

- producing a polarized laser read beam;
- producing from said read beam a pair of focussed cross-polarized component beams;
- focussing said component beam pair at adjacent locations in a medium containing inhomogeneities, and

- scanning said focussed beams through said medium to produce an interference image of inhomogeneities in said medium.

8. A method of writing and subsequently reading optical data with three-dimensional resolution, comprising:

- providing a three-dimensional medium of photosensitive material capable of modification by the application of light of a predetermined intensity;

- producing a single, intense beam of light;

- focussing said beam of light at a focal point within said medium;

- scanning said focal point through said medium to produce at selected points in said medium two-photon excitation of said medium to produce a modification of a selected characteristic of the medium, each such modification representing an optical data bit in said medium; and

thereafter reading optical data bits produced in said medium by scanning a focused reading beam through said medium to produce an interference pattern corresponding to said optical data bits.

1/4

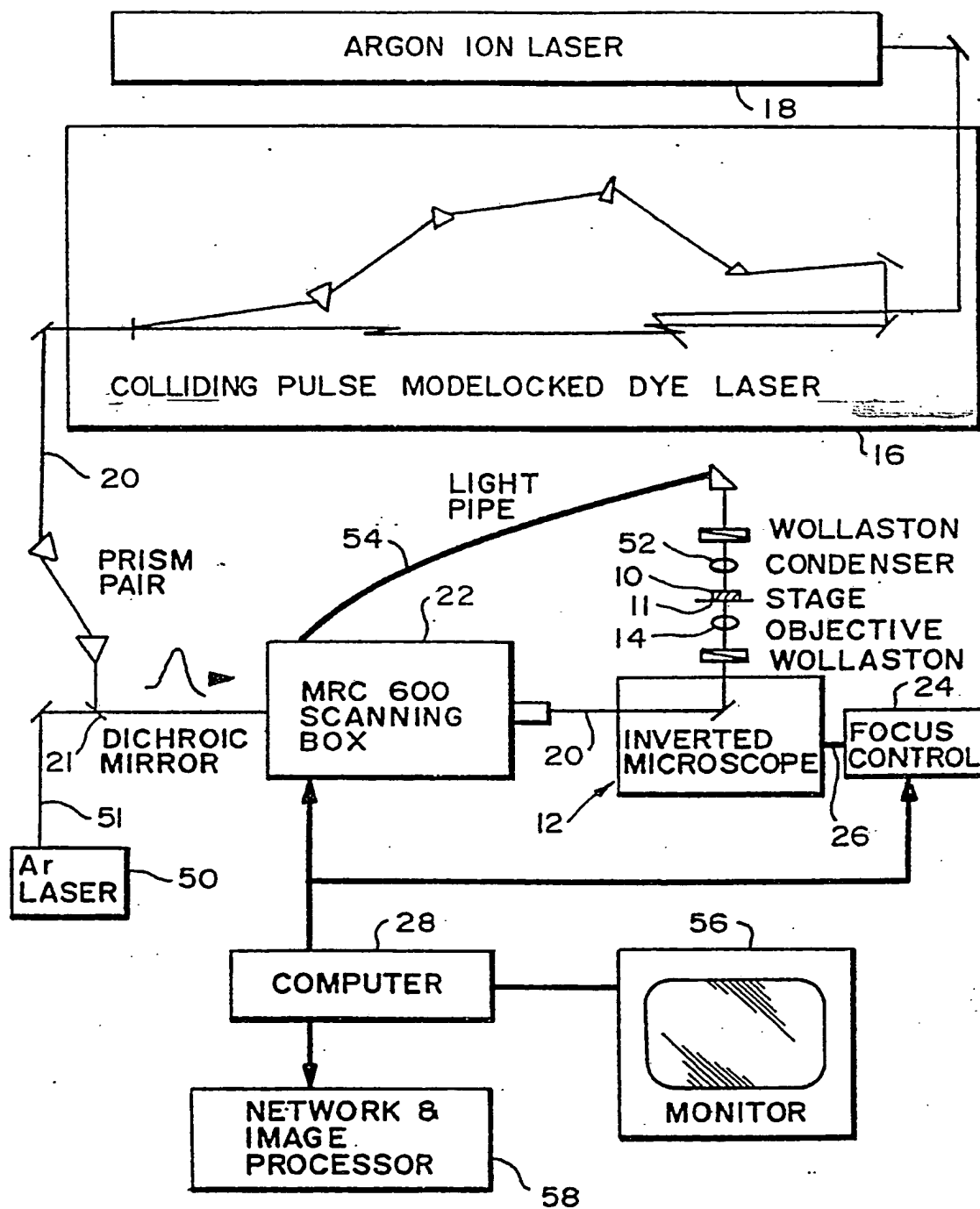


FIG. 1

2/4

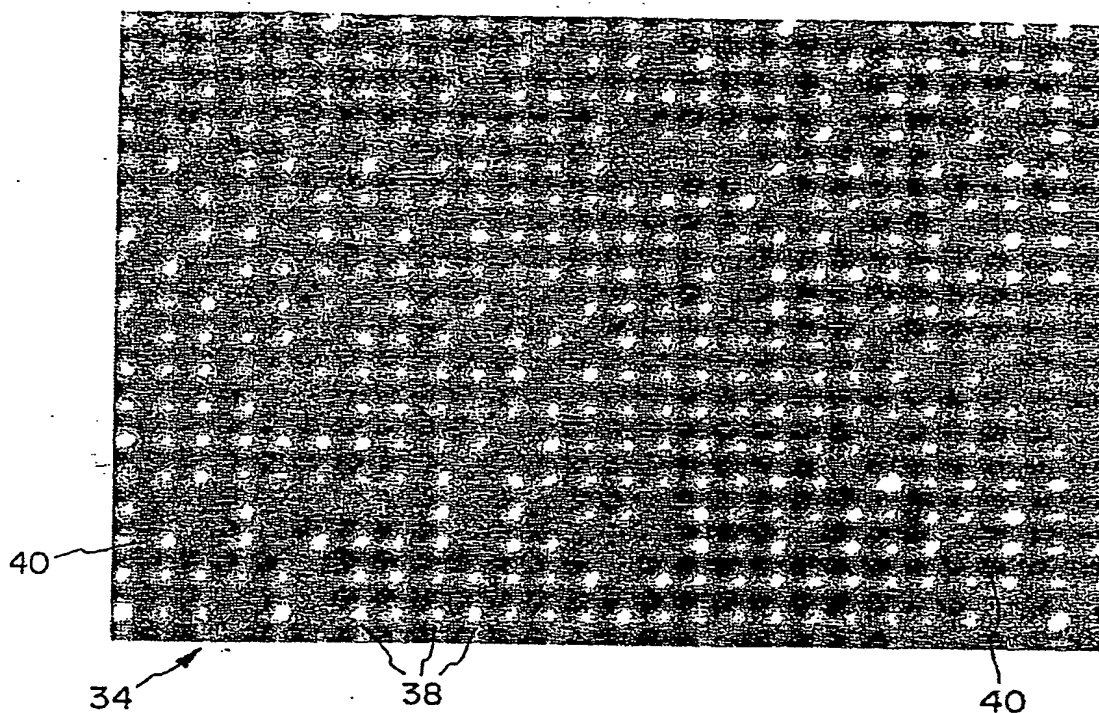


FIG. 2

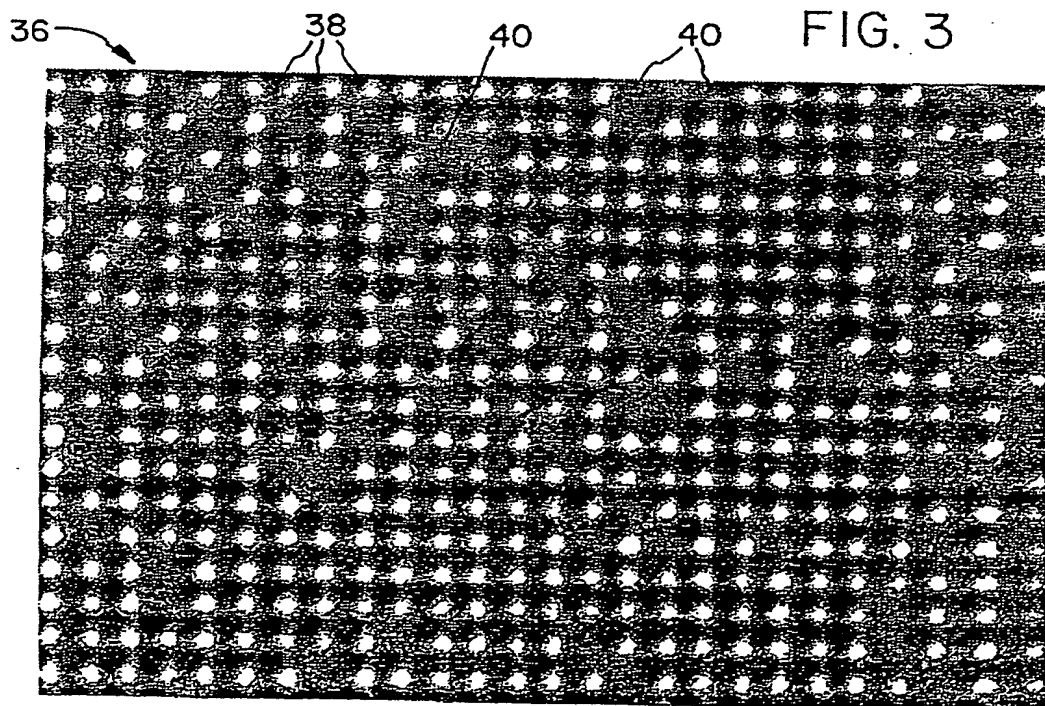


FIG. 3

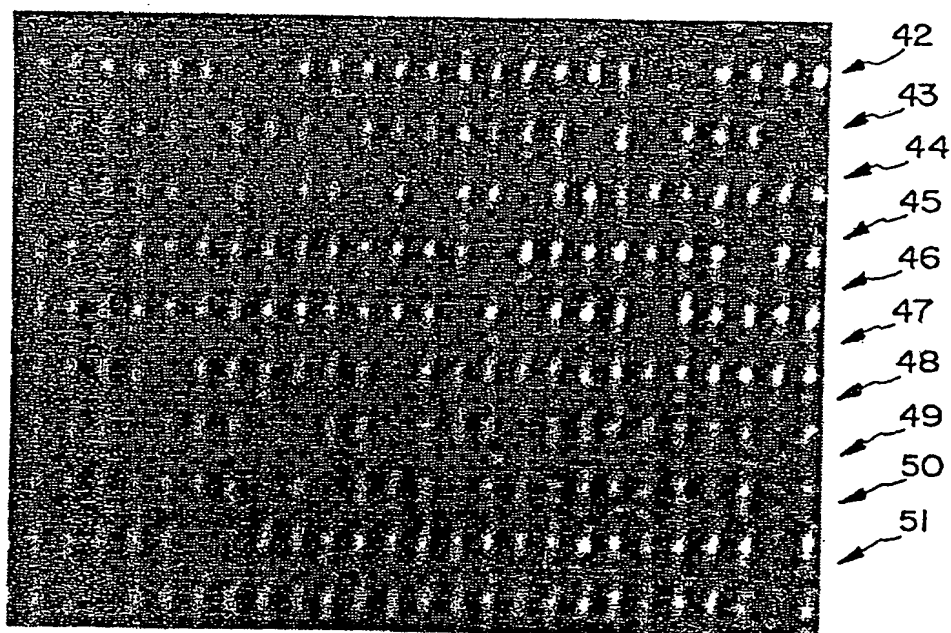


FIG. 4

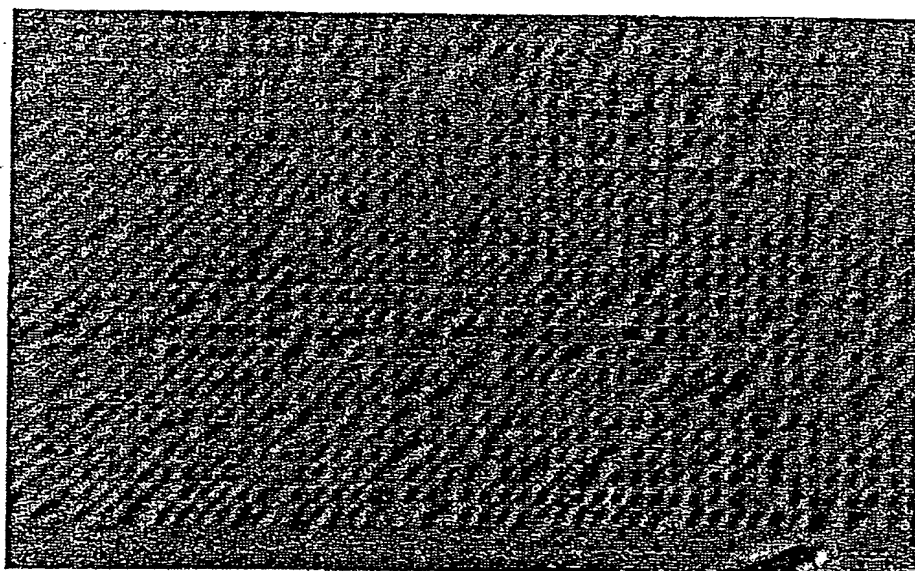


FIG. 5

INTERNATIONAL SEARCH REPORT

 International application No.
 PCT/US92/05858

A. CLASSIFICATION OF SUBJECT MATTER IPC(5) : G11C 13/04 US CL : 364/127,107 According to International Patent Classification (IPC) or to both national classification and IPC		
B. FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) U.S. : 364/106,107,120,127 Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)		
C. DOCUMENTS CONSIDERED TO BE RELEVANT		
Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US,A, 4,333,165 (SWAINSON ET AL) 01 JUNE 1982 COLUMN 13, LINES 12 TO 24	1-6
X	US,A, 4,288,862 (SWAINSON ET AL) 08 SEPTEMBER 1981 SEE COLUMN 28, LINE 46 TO COLUMN 29, LINE 10.	7-8
<input type="checkbox"/> Further documents are listed in the continuation of Box C. <input type="checkbox"/> See patent family annex.		
* Special categories of cited documents: "A" document defining the general state of the art which is not considered to be part of particular relevance "E" earlier document published on or after the international filing date "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified) "O" document referring to an oral disclosure, use, exhibition or other means "P" document published prior to the international filing date but later than the priority date claimed "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art "Z" document member of the same patent family		
Date of the actual completion of the international search 09 SEPTEMBER 1992		Date of mailing of the international search report 24 OCT 1992
Name and mailing address of the ISA/ Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231 Facsimile No. NOT APPLICABLE		Authorized officer JOSEPH A. POPEK Telephone No. (703) 308-2806